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**Study of newly discovered two dimensional cobalt based
perovskite compounds doped with various rare earth elements**

A thesis submitted in fulfillment of the requirement for the award of the degree

Doctor of Philosophy (PhD)

from

University of Wollongong

by

Qi Wen Yao,

B.S. (UNSW)

M.E. (Mat. Eng. UOW)

Institute for Superconducting and Electronic Materials

Faculty of Engineering

2008

Declaration

I, Qi Wen Yao, declare that this thesis, submitted in fulfilment of the requirements for the award of Doctor of Philosophy, in the Institute for Superconducting and Electronic Materials, in the Faculty of Engineering, University of Wollongong, is wholly original work unless otherwise referenced or acknowledged. This thesis has not been submitted for qualifications at any other academic institution.

Qi Wen Yao

Wollongong

November 2008

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ABSTRACT

This thesis focuses on the study of a newly discovered two-dimensional CoO_2 layer structured perovskite compound, Sr_2CoO_4 . To explore doping effects on the physical properties of the new compound, a systematic and detailed experimental study has been carried out, relating to the aspects of synthesis, structure, transport and magneto-transport behaviour, magnetism, and dielectricity. Theoretical investigations have also been carried out for both crystal and electronic band structures, using Rietveld refinement and first-principles band structure calculations.

Various rare earth element doped Sr_2CoO_4 polycrystalline compounds ($\text{Sr}_{2-x}\text{RE}_x\text{CoO}_4$, where $x = 0.25 - 1.5$ and $\text{RE} = \text{Pr}, \text{La}, \text{Gd}, \text{Eu}, \text{and Nd}$) were studied systematically. It has been found that the size and valence, as well as the doping level of the rare elements, control the physical properties of the Sr_2CoO_4 . Ferromagnetic behaviour is found to exist in some of the doped compounds and to have interesting properties. It was observed that the lattice parameter a remains relatively stable, with values around 3.7 to 3.8 Å for $x = 1$ for various RE doped compounds of $\text{Sr}_{2-x}\text{RE}_x\text{CoO}_4$. The Co-O(2) in-plane bond lengths were also not sensitive to different RE dopants for the $x = 1$ doping level. In contrast, lattice parameter c , as well as the out-of-plane bond length Co-O(1), varies with different RE dopants in the compound for the doping level of $x = 1$, and consequently, the unit cell volume also changes, depending on the RE dopant. It was found that Sr_2CoO_4 is the most tolerant to Pr as a dopant. For x values from 0.5 to 1.5, single phase Sr_2CoO_4 structured samples were achieved. La doping was compatible with single phase compounds for $x = 1 - 1.25$, and for Eu doping, single phase compounds

were formed for $x = 0.75 - 1$. For both Gd and Nd doping, near single phases are formed for all the tested x values ($x = 0.5 - 1.25$),

Our results show that some of the Pr-doped and La-doped single phase samples have large coercive fields, and hence, they have good potential industrial applications (permanent magnets in electric motors, magnetic recording media, etc.), while Eu-doped samples can have high magnetoresistance (MR) values, making the Eu-doped compound a good candidate for application as a colossal magnetic resistance (CMR) material.

For the Pr-doped $\text{Sr}_{2-x}\text{Pr}_x\text{CoO}_4$, the lattice parameter c decreased with the Pr doping level x . The Curie temperature (T_C) was found to be 200 K for $\text{Sr}_{1.5}\text{Pr}_{0.5}\text{CoO}_4$. The resistivities were found to increase with doping level x . A large coercive field of about 1 Tesla (T) was found for the sample with $x = 0.75$. The values of the dielectric constant (ϵ) were over 2000 at low frequencies of less than 1 kHz (not shown here) and gradually decreased with increasing frequencies. The ϵ of the $x = 1$ sample is greater than that of the $x = 0.75$ sample, indicating that the charge induced capacitance in the $x = 1$ sample is greater than that of the $x = 0.75$ sample. This is in agreement with the trend of their resistivity measurements.

For the Eu-doped compounds of Sr_2CoO_4 , single 214 phase was achieved for $x = 0.75 - 1$. The lattice parameter c decreased with the Eu doping level. The Curie temperatures were found to be around 160-200 K for samples with $x = 0.75$ and 1. An antiferromagnetic transition is observed at 35 K for the $x = 0.75$ sample. Magnetic

semiconductor characteristics were observed for all the Eu-doped samples. The existence of unusually high magnetoresistance in these compounds makes them stand out from the rest of the RE doped compounds in this regard. For example, the sample with $x = 1.25$ showed a MR value of about 46% at 8 Tesla at 100 K.

For the La-doped Sr_2CoO_4 compounds, the temperature dependence of the resistivity shows a semiconductor-like behavior over a wide range of temperatures, a metal-insulator transition at 240 K, and an upturn at 160 K for the $x = 1$, 1.25, and 0.75 samples. The coercive field was about 1 T for the sample with $x = 0.75$, while it is about 0.05 T for the $x = 0.75$ and 0.1 T for the $x = 1.25$ sample. A negative field hysteresis in the magnetoresistance in close correlation with the coercive field has been observed and can be explained by the grain boundary tunneling effect. First-principles band structure calculations were carried out for $\text{Sr}_{1.5}\text{La}_{0.5}\text{CoO}_4$, and the results indicate that the system is metallic, with a high spin polarization which is responsible for the observed large magnetoresistance. The phonon density of states (PDOS) reveals that the Co 3d electrons and planar oxygen electrons are responsible for the high spin polarization at the Fermi surface in the compound.

The Gd-doped Sr_2CoO_4 compounds were found to be paramagnetic semiconductors with MR values of only around 3 to 5%. Their transport properties can be described by the hopping model for semiconductors. Band structure calculations indicate that the spin polarization is high in the Gd-doped Sr_2CoO_4 .

Nd-doped compounds were found to be ferromagnetic semiconductors at temperatures of about 250 and 170 K for the $x = 1$ and 0.75 samples, respectively. Their MR values were about 5 %, similar to those of the Gd-doped samples. The Nd doping raised the Curie temperature of Sr_2CoO_4 , so that it reached 210 K for doping with Nd at levels up to $x = 0.5$.

Table of Contents

Introduction		1
Chapter 1	Fundamentals and Literature Review	11
	1. 1 Fundamentals	11
	1.1.1 Magnetic material category	11
	1.1.2 Colossal magnetoresistance (CMR)	13
	1.1.3 Giant Magnetoimpedance (GMI)	14
	1.1.4 Interlayer coupling	17
	1.1.5 Tunneling magnetoresistance	18
	1.1.6. Ferromagnetism and its application	19
	1.1.7. Curie Weiss Law	21
	1.1.8. Variable Range Hopping (VHR) - - the Mott Theory	22
	1.2 Relevant research - K ₂ NiF ₄ -type compounds	23
Chapter 2	Experimental procedures and techniques	53
	2.1. Sample preparation	53
	2.2 Sample characterisation	56
	2.3 The Physical Property Measurement System (PPMS)	56
	2.4 The Magnetic Property Measurement System (MPMS)	58
	2.5 Dielectric measurements	59
Chapter 3	Pr doped Sr ₂ CoO ₄ (Sr _{2-x} Pr _x CoO ₄)	61

	3.1 Introduction	61
	3.2 Structural properties	62
	3.4 Transport properties	73
	3.5 Magnetisation properties	75
	3.6 Dielectric properties	79
	3.7 Summary	81
Chapter 4	La doped Sr_2CoO_4 ($\text{Sr}_{2-x}\text{La}_x\text{CoO}_4$, $x = 0.5, 0.75, 1, 1.25$)	82
	4.1 Introduction	82
	4.2 Structural properties	83
	4.3 Transport properties	90
	4.4 Magnetisation properties	92
	4.5 Magnetoresistance	95
	4.6 Band structure	97
	4.7 Summary	98
Chapter 5	Eu doped Sr_2CoO_4 ($\text{Sr}_{2-x}\text{Eu}_x\text{CoO}_4$, $x = 0.25, 0.75, 1, 1.25$)	100
	5.1 Introduction	100
	5.2 Structural properties	100
	5.3 Transport properties	106
	5.4 Magnetisation properties	108
	5.5 Magnetoresistance	109
	5.6 Summary	111
Chapter 6	Gd doped Sr_2CoO_4 ($\text{Sr}_{2-x}\text{Gd}_x\text{CoO}_4$, $x = 0.5, 0.75, 1, 1.25$)	112

6.1 Introduction	112
6.2 Structural properties	112
6.4 Magnetisation properties	122
6.5 Magnetoresistance	125
6.6 Summary	129
Chapter 7 Nd doped Sr_2CoO_4 ($\text{Sr}_{2-x}\text{Nd}_x\text{CoO}_4$, $x = 0.5, 0.75, 1, 1.25$)	130
7.1 Introduction	130
7.2 Structural properties	130
7.3 Transport properties	138
7.4 Magnetisation properties	140
7.5 Magnetoresistance	143
7.6 Summary.	145
CHAPTER 8 Summary for various doping compounds	146
Publication List	149
References	152

List of Figures

Fig. 1	Structural views of $\text{Na}_{0.7}\text{CoO}_2$ (left) and $\text{Na}_x\text{CoO}_2 \cdot y\text{H}_2\text{O}$ (right) [32].
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Fig. 2	Crystal structure of Sr_2CoO_4 .	6
Fig. 3	Temperature dependence of the resistivity (for the $\text{Sr}_{2-y}\text{Y}_y\text{CoO}_4$ system. Inset shows the activation energy, E_a (reproduced from [1]).	7
Fig. 4	Temperature dependence of the field cooled dc magnetization, measured at a magnetic field of 20 Oe (reproduced from [1]).	8
Fig. 5	Magnetic hysteresis loops for $\text{Sr}_{2-y}\text{Y}_y\text{CoO}_4$ (upper panel) and field-hysteretic magnetoresistance (lower panel) for Sr_2CoO_4 at 5 K (reproduced from [1]).	9
Fig. 1. 1	Schematic illustration of the behaviour of the exchange coupling vs distance	17
Fig. 1. 2	Illustration of tunneling magnetoresistance (TMR). Two ferromagnetic layers separated by an insulating layer (i = electron current)(reproduced from [41]).	19
Fig. 1. 3	Schematic representation of the K_2NiF_4 structure displayed by the $n = 1$ RP phases A_2BO_4 . Fig. reproduced from [33]	25
Fig. 1. 4	dc magnetization vs temperature for $(\text{SrO})(\text{SrCoO}_3)_n$ where $n = 1, 2, 3, 4$ and ∞ . Inset is Curie–Weiss fitting form $n=1$ sample [90].	26
Fig. 1. 5	Magnetic hysteresis loops (lower panel) and field-hysteretic magnetoresistance	30
Fig. 1. 6	Temperature dependence of the magnetization M for Sr_2CoO_4 thin films. Inset shows field dependence of the magnetization M measured at 5 K. Fig. reproduced from [92].	31

Fig. 1. 7	The relationship between the reciprocal of the magnetic susceptibility($1/\chi$) of $\text{Ca}_{1+x}\text{Sm}_{1-x}\text{CoO}_4$ and temperature (T). Fig. reproduced from [93].	33
Fig. 1. 8	Temperature dependence of magnetoresistance of polycrystalline SrLaCoO_4 at 2.5 and 5 T field [21]. Reproduced from [94].	35
Fig. 1. 9	Temperature dependence of field cooled dc magnetization (reproduced from [125]).	36
Fig. 1. 10	Temperature dependence of electric resistivity for the $\text{Sr}_{2-x}\text{Ho}_x\text{CoO}_4$ system (reproduced from [125]).	37
Fig. 1. 11	Temperature dependence of magnetoresistance (MR) for the $\text{Sr}_{2-x}\text{Ho}_x\text{CoO}_4$ system under 7 T field (reproduced from [125]).	38
Fig. 1. 12	Magnetoresistance (upper panel) and magnetization M (lower panel) for pure and Y or Gd doped Sr_2CoO_4 .	39
Fig. 1. 13	Temperature dependence of the field-cooled magnetization for both Gd and Y doped Sr_2CoO_4 .	41
Fig.1. 14	Magnetic hysteresis loops (lower panel) and field-hysteretic magnetoresistance (upper panel) for Sr_2CoO_4 at 5 K [140].	42
Fig.1. 15	Temperature dependence of the magnetization M for Sr_2CoO_4 thin films. Inset shows the field dependence of the magnetization M measured at 5 K [140].	43
Fig.1. 16	The temperature dependence of the reciprocal of the magnetic susceptibility ($1/\chi$) of $\text{Ca}_{1+x}\text{Sm}_{1-x}\text{CoO}_4$ [142].	45
Fig.1. 17	Temperature dependence of the magnetoresistance of polycrystalline SrLaCoO_4 for 2.5 T and 5 T fields [21]. Reproduced from [143].	46

Fig.1. 18	Temperature dependence of the field-cooled dc magnetization of $\text{Sr}_{2-x}\text{Ho}_x\text{CoO}_4$ [144].	47
Fig.1. 19	Temperature dependence of the electrical resistivity for the $\text{Sr}_{2-x}\text{Ho}_x\text{CoO}_4$ system [144].	48
Fig.1. 20	Temperature dependence of the magnetoresistance (MR) for the $\text{Sr}_{2-x}\text{Ho}_x\text{CoO}_4$ system under a 7 T field [144].	48
Fig.1. 21	Magnetoresistance (upper panel) and magnetization M (lower panel) for pure and Y or Gd doped Sr_2CoO_4 [1].	48
Fig.1. 22	Magnetoresistance (upper panel) and magnetization M (lower panel) for pure and Y or Gd doped Sr_2CoO_4 [1].	50
Fig. 2. 1	Sample preparation flow chart.	55
Fig. 3. 1	XRD patterns for $\text{Sr}_{2-x}\text{Pr}_x\text{CoO}_4$ where $x = 0.25-1.5$.	62
Fig. 3. 2	Crystal structure of Sr_2CoO_4 .	63
Fig. 3. 3	Rietveld refinement for $\text{Sr}_{1.5}\text{Pr}_{0.5}\text{CoO}_4$.	64
Fig. 3. 4	Rietveld refinement for $\text{Sr}_{1.25}\text{Pr}_{0.75}\text{CoO}_4$.	65
Fig. 3. 5	Rietveld refinement for SrPrCoO_4 .	66
Fig. 3.6	Rietveld refinement for $\text{Sr}_{0.75}\text{Pr}_{1.25}\text{CoO}_4$.	66
Fig. 3.7	Rietveld refinement for $\text{Sr}_{0.5}\text{Pr}_{1.5}\text{CoO}_4$.	67
Fig. 3. 8	The lattice parameter a vs x for $\text{Sr}_{2-x}\text{Pr}_x\text{CoO}_4$.	70
Fig. 3. 9	The lattice parameter c vs x for $\text{Sr}_{2-x}\text{Pr}_x\text{CoO}_4$.	71
Fig. 3. 10	The Co-O bond length vs x for $\text{Sr}_{2-x}\text{Pr}_x\text{CoO}_4$.	72
Fig. 3. 11	The unit cell volume vs x for $\text{Sr}_{2-x}\text{Pr}_x\text{CoO}_4$.	72
Fig. 3. 12	Resistance vs temperature for $\text{Sr}_{1.25}\text{Pr}_{0.75}\text{CoO}_4$.	74

Fig. 3. 13	ρ vs $\exp(1/T)^{1/3}$ for $\text{Sr}_{1.25}\text{Pr}_{0.75}\text{CoO}_4$. Straight lines are linear fitting to the 2D VRH model.	75
Fig. 3. 14	The temperature dependence from 5 K to 300 K of the magnetization of the $\text{Sr}_{2-x}\text{Pr}_x\text{CoO}_4$ samples with $x = 0.25, 0.5, 0.75, 1, 1.25$ measured in a magnetic field of 0.2 Tesla.	76
Fig. 3. 15	Temperature dependence of the $\text{Sr}_{2-x}\text{Pr}_x\text{CoO}_4$ samples' magnetization in low temperature range.	77
Fig. 3. 16	Temperature dependence of the inverse molar susceptibility, $(\chi-1)$ for $\text{Sr}_{2-x}\text{Pr}_x\text{CoO}_4$, measured at a magnetic field of 2 kOe.	78
Fig. 3. 17	Magnetic hysteresis loops for $\text{Pr}_x\text{Sr}_{2-x}\text{CoO}_4$.	79
Fig. 3. 18	Frequency dependence of dielectric constant for $\text{Sr}_{2-x}\text{Pr}_x\text{CoO}_4$ ($x = 1, 1.25$) at room temperature.	80
Fig. 3. 19	Frequency dependence of dielectric loss for $\text{Sr}_{2-x}\text{Pr}_x\text{CoO}_4$ ($x = 1, 1.25$) at room temperature.	81
Fig. 4. 1	XRD patterns of $\text{Sr}_{2-x}\text{La}_x\text{CoO}_4$ with $x = 0.75, 1, 1.25$	84
Fig. 4. 2	Rietveld refinement for $\text{Sr}_{1.25}\text{La}_{0.75}\text{CoO}_4$. The observed (crosses), calculated (solid line) and difference (bottom line) profiles at 300K for the $x=0.75$ sample (refinement factors $R_p=14.8\%$, $R_b=2.9\%$; blue-strip range was ignored in refinement).	85
Fig. 4. 3	Rietveld refinement for SrLaCoO_4 . The observed (crosses), calculated (solid line), and difference (bottom line) profiles at 300K for the $x = 1$ sample (refinement factors $R_p=12.58\%$, $R_b=7.3\%$).	86
Fig. 4. 4	Rietveld refinement for $\text{Sr}_{0.75}\text{La}_{1.25}\text{CoO}_4$. The observed (crosses), calculated (solid line) and difference (bottom line) profiles at 300K (refinement factors $R_p=12.58\%$, $R_b=7.3\%$ for the $x = 1.25$ sample).	87

Fig. 4. 5	Co-O bond lengths vs. doping level x .	90
Fig. 4. 6	The temperature dependence of the electrical resistivity (ρ) of the $\text{Sr}_{2-x}\text{La}_x\text{CoO}_4$ samples with $x = 0.75, 1, 1.25$, from 5 K to 350 K, measured in zero field.	91
Fig. 4. 7	$\ln(\rho)$ vs. $\exp(1/T)^{1/4}$ for $\text{Sr}_{2-x}\text{La}_x\text{CoO}_4$. Straight lines are linear fittings to the 2D VRH model.	92
Fig. 4. 8	The temperature dependence from 5 K to 350 K of the magnetization of the $\text{Sr}_{2-x}\text{La}_x\text{CoO}_4$ samples with $x = 0.75, 1, 1.25$, measured in a magnetic field of 0.2 Tesla.	94
Fig. 4. 9	Temperature dependence of the inverse susceptibility (χ^{-1}) for the $\text{Sr}_{2-x}\text{La}_x\text{CoO}_4$ samples, measured at a magnetic field of 2000 Oe.	94
Fig. 4. 10	Magnetization hysteresis loops for the $\text{Sr}_{2-x}\text{La}_x\text{CoO}_4$ samples with $x = 0.75, 1$, and 1.25 , measured at 10 K.	95
Fig. 4. 11	Field hysteretic magnetoresistance for $\text{Sr}_{1.25}\text{La}_{0.75}\text{CoO}_4$ at 150 K.	96
Fig. 4. 12	Magnetoresistance hysteresis for $\text{Sr}_{0.75}\text{La}_{1.25}\text{CoO}_4$ at 5 K.	96
Fig. 4. 13	Rate of change of the resistivity for the sample with $x = 1.25$ at various temperatures in magnetic field.	97
Fig. 4. 14	Calculated total density of states of $\text{Sr}_{1.5}\text{La}_{0.5}\text{CoO}_4$ compound and the partial density of states of Co 3d (upper panel); and the partial density of states of planar and apical oxygen (lower panel), with the Fermi energy set at zero.	98
Fig. 5. 1	XRD patterns of the $\text{Sr}_{2-x}\text{Eu}_x\text{CoO}_4$ samples with $x = 0.25, 0.75, 1, 1.25$, measured at room temperature.	101

Fig. 5. 2	Rietveld refinement for $\text{Sr}_{1.25}\text{Eu}_{0.75}\text{CoO}_4$ showing the observed (crosses), calculated (solid line) and difference (bottom line) profiles at 300K (refinement factors $R_p=11.4\%$, $R_b=2.9\%$).	102
Fig. 5. 3	Rietveld refinement for SrEuCoO_4 showing the observed (crosses), calculated (solid line) and difference (bottom line) profiles at 300K (refinement factors $R_p=13.4\%$, $R_b=2.1\%$; blue-strip range was ignored in refinement).	102
Fig. 5. 4	Rietveld refinement for $\text{Sr}_{0.75}\text{Eu}_{1.25}\text{CoO}_4$ showing the observed (crosses), calculated (solid line) and difference (bottom line) profiles at 300K (refinement factors $R_p=13.8\%$, $R_b=4.6\%$; blue-strip ranges were ignored in refinement).	103
Fig. 5. 5	Co-O bond lengths vs. doping level x .	106
Fig. 5. 6	Temperature dependence of the resistivity of $\text{Sr}_{2-x}\text{Eu}_x\text{CoO}_4$ with $x = 0.75, 1, 1.25$.	107
Fig. 5. 7	$\ln(\rho)$ vs. $\exp(1/T)^{1/3}$ for $\text{Sr}_{2-x}\text{Eu}_x\text{CoO}_4$ with $x = 0.75, 1, 1.25$. Straight lines are linear fittings to the 2D VRH model.	108
Fig. 5. 8	Field cooled and zero field cooled temperature dependence of magnetization for $\text{Sr}_{2-x}\text{Eu}_x\text{CoO}_4$ with $x = 0.75$ and 1 .	109
Fig. 5. 9	Magnetoresistance hysteresis for the $\text{Sr}_{1.25}\text{Eu}_{0.75}\text{CoO}_4$ sample at 100 K.	110
Fig. 5. 10	Magnetoresistance hysteresis for the SrEuCoO_4 sample at 100 K.	110
Fig. 5. 11	Magnetoresistance hysteresis for the $\text{Sr}_{0.75}\text{Eu}_{1.25}\text{CoO}_4$ sample at 100 K.	111

Fig. 6. 1	XRD patterns of $\text{Sr}_{2-x}\text{Gd}_x\text{CoO}_4$ with $x = 0.5, 0.75, 1, 1.25$ (* indicates unknown impurity).	113
Fig. 6. 2	Rietveld refinement for $\text{Sr}_{1.5}\text{Gd}_{0.5}\text{CoO}_4$ showing the observed (crosses), calculated (solid line) and difference (bottom line) profiles at 300K (refinement factors $R_p=14.5\%$, $R_b=5.6\%$; blue-strip ranges were ignored in refinement).	114
Fig. 6. 3	Rietveld refinement for $\text{Sr}_{1.25}\text{Gd}_{0.75}\text{CoO}_4$ showing the observed (crosses), calculated (solid line) and difference (bottom line) profiles at 300K (refinement factors $R_p=14.3\%$, $R_b=6.2\%$; blue-strip range was ignored in refinement).	114
Fig. 6. 4	Rietveld refinement for SrGdCoO_4 showing the observed (crosses), calculated (solid line) and difference (bottom line) profiles at 300K (refinement factors $R_p=12.0\%$, $R_b=3.2\%$; blue-strip range was ignored in refinement).	115
Fig. 6. 5	Rietveld refinement for $\text{Sr}_{0.75}\text{Gd}_{1.25}\text{CoO}_4$ showing the observed (crosses), calculated (solid line) and difference (bottom line) profiles at 300K (refinement factors $R_p=16.6\%$, $R_b=7.6\%$; blue-strip range was ignored in refinement).	115
Fig. 6. 6	Lattice parameters a and c vs. x for $\text{Sr}_{2-x}\text{Gd}_x\text{CoO}_4$.	119
Fig. 6. 7	Co-O bond lengths vs. doping level x .	120
Fig. 6. 8	Temperature dependence of the resistivity of $\text{Sr}_{2-x}\text{Gd}_x\text{CoO}_4$ with $x = 0.75, 1, 1.25$.	121
Fig. 6. 9	$\ln(\rho)$ vs $\exp(1/T)^{1/3}$ for $\text{Sr}_{2-x}\text{Gd}_x\text{CoO}_4$ with $x = 0.75, 1, 1.25$. Straight lines are linear fittings to the 2D VRH model.	122

Fig. 6. 10	Temperature dependence of the magnetization for $\text{Sr}_{2-x}\text{Gd}_x\text{CoO}_4$ with $x = 0.75$ and 1 .	123
Fig. 6. 11	Temperature dependence of the inverse susceptibility (χ^{-1}) for $\text{Sr}_{2-x}\text{Gd}_x\text{CoO}_4$, measured at a magnetic field of 2000 Oe.	124
Fig. 6. 12	Magnetization hysteresis measurements for $\text{Sr}_{2-x}\text{Gd}_x\text{CoO}_4$ with $x = 0.75$ and 1 , measured at 10 K.	124
Fig. 6. 13	Magnetoresistance hysteresis for $\text{Sr}_{1.25}\text{Gd}_{0.75}\text{CoO}_4$ at 100 K.	125
Fig. 6. 14	Magnetoresistance hysteresis for SrGdCoO_4 at 100 K.	126
Fig. 6. 15	The band structure (upper panel) and density of states (lower panel) of the $\text{Sr}_{1.5}\text{Gd}_{0.5}\text{CoO}_4$ sample.	128
Fig. 6. 16	Partial density of states of Co in the $\text{Sr}_{1.5}\text{Gd}_{0.5}\text{CoO}_4$ sample.	128
Fig. 7.1	XRD patterns of $\text{Sr}_{2-x}\text{Nd}_x\text{CoO}_4$ with $x = 0.5, 0.75, 1, 1.25$.	131
Fig. 7.2	Rietveld refinement for $\text{Sr}_{1.5}\text{Nd}_{0.5}\text{CoO}_4$ showing the observed (crosses), calculated (solid line) and difference (bottom line) profiles at 300K for the $x = 0.5$ sample (refinement factors $R_p=14.9\%$, $R_b=7.4\%$; blue-strip ranges were ignored in refinement).	132
Fig. 7.3	Rietveld refinement for $\text{Sr}_{1.25}\text{Nd}_{0.75}\text{CoO}_4$ showing the observed (crosses), calculated (solid line) and difference (bottom line) profiles at 300K for the $x = 0.75$ sample (refinement factors $R_p=18.4\%$, $R_b=11.5\%$; blue-strip ranges were ignored in refinement).	132
Fig. 7.4	Rietveld refinement for SrNdCoO_4 showing the observed (crosses), calculated (solid line) and difference (bottom line) profiles at 300K for the $x = 1$ sample (refinement factors $R_p=10.9\%$, $R_b=6.4\%$; blue-strip range was ignored in refinement).	133

Fig. 7.5	Rietveld refinement for $\text{Sr}_{0.75}\text{Nd}_{1.25}\text{CoO}_4$ showing the observed (crosses), calculated (solid line) and difference (bottom line) profiles at 300K for the $x = 1$ sample (refinement factors $R_p=13.9\%$, $R_b=8.4\%$).	133
Fig. 7.6	Co-O bond lengths v.s doping level x .	137
Fig. 7.7	Lattice parameters a and c vs. x for $\text{Sr}_{2-x}\text{Nd}_x\text{CoO}_4$.	137
Fig. 7.8	The unit cell volume vs. x for $\text{Sr}_{2-x}\text{Nd}_x\text{CoO}_4$.	138
Fig. 7.9	Resistance vs. temperature for $\text{Sr}_{2-x}\text{Nd}_x\text{CoO}_4$ ($x = 0.75, 1$).	139
Fig. 7.10	$\ln(\rho)$ vs. $\exp(1/T)^{1/3}$ for $\text{Sr}_{2-x}\text{Nd}_x\text{CoO}_4$ ($x = 0.75, 1$). Straight lines are linear fittings to the 2D VRH model.	140
Fig. 7.11	The temperature dependence from 10 K to 340 K of the magnetization of the $\text{Sr}_{2-x}\text{Nd}_x\text{CoO}_4$ samples with $x = 0.75, 1$, measured in a magnetic field of 0.2 Tesla.	141
Fig. 7.12	Temperature dependence of the inverse susceptibility (χ^{-1}) for $\text{Sr}_{2-x}\text{Nd}_x\text{CoO}_4$, measured at a magnetic field of 2000 Oe.	142
Fig. 7.13	Magnetic hysteresis loops for $\text{Sr}_{2-x}\text{Nd}_x\text{CoO}_4$ with $x = 0.75, 1$.	143
Fig. 7.14	Magnetoresistance hysteresis for SrGdCoO_4 at 100 K.	144
Fig. 7.15	Magnetoresistance hysteresis for $\text{Sr}_{1.25}\text{Nd}_{0.75}\text{CoO}_4$ at 100 K.	144

List of Tables

Table 1. 1	Summary of different types of magnetic behaviour (Source: University of Birmingham website).	11
Table 2. 1	Single phase polycrystalline samples $\text{Sr}_{2-x}\text{RE}_x\text{CoO}_4$ that have been attempted to produce.	53
Table 3. 1	Crystal data of $\text{Sr}_{2-x}\text{Pr}_x\text{CoO}_4$ ($x=0.5, 0.75, 1, 1.25$) - Space group: $I4/mmm$.	67
Table 4. 1	Crystal data of $\text{Sr}_{2-x}\text{La}_x\text{CoO}_4$ ($0.75, 1, 1.25$) - Space group: $I4/mmm$.	88
Table 4. 2	Structural parameters of $\text{Sr}_{2-x}\text{La}_x\text{CoO}_4$ with $x = 0.75, 1$ & 1.25 from the Rietveld refinement (space group $I4/mmm$) results.	89
Table 5. 1	Crystal data of $\text{Sr}_{2-x}\text{Eu}_x\text{CoO}_4$ ($x = 0.75, 1, 1.25$) - Space group: $I4/mmm$.	104
Table 5. 2	Structural parameters of $\text{Sr}_{2-x}\text{Eu}_x\text{CoO}_4$ with $x = 0.75, 1$ & 1.25 from the Rietveld refinement (space group $I4/mmm$) results.	105
Table 6. 1	Crystal data of $\text{Sr}_{2-x}\text{Gd}_x\text{CoO}_4$ ($x = 0.5, 0.75, 1, 1.25$) - Space group: $I4/mmm$.	116
Table 6. 2	Structural parameters of $\text{Sr}_{2-x}\text{Gd}_x\text{CoO}_4$ with $x = 0.5, 0.75, 1, 1.25$ from the Rietveld refinement (space group $I4/mmm$) results.	118
Table 7. 1	Crystal data of $\text{Sr}_{2-x}\text{Nd}_x\text{CoO}_4$ ($x = 0.5, 0.75, 1, 1.25$) - Space group: $I4/mmm$.	134
Table 7. 2	Structural parameters of $\text{Sr}_{2-x}\text{Nd}_x\text{CoO}_4$ with $x = 0.5, 0.75, 1, 1.25$ from the Rietveld refinement (space group $I4/mmm$) results.	136

Table 8. 1 Summarization of the properties for different doped compounds.

146